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Operated By
CARBIDE AND CARBON CHEMICALS COMPANY

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SUBJECT: Summary of Efforts to Recover 235 Spillage at 7500 Area

TO: C. E. Winters

FROM: S. E. Beall

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INTER-COMPANY CORRESPONDENCE

OAK RIDGE NATIONAL LABORATORY
Operated By
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TO C. E. Winters
LOCATION Bldg. 9204-1, Y-12

DATE September 10, 1952
ANSWERING LETTER DATE

ATTENTION
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SUBJECT Summary of Efforts to Recover 235 Spillage at 7500 Area

Since the loss of approximately 800 grams of U^{235} from the HRE in mid-July, an extensive effort has been made to locate and recover the spilled material. This memorandum will summarize the results of the recovery effort.

A brief description of the portion of the fuel system from which the loss occurred will aid in understanding the recovery problem. Oxygen gas, necessary to minimize corrosion, is fed continuously into the fuel system through a $\frac{1}{4}$" stainless steel line which originates at an oxygen cylinder station outside the reactor building. The line passes through the control room, where the cylinder pressure is reduced by high pressure gas regulators, and enters the reactor shield structure at the cell provided for heavy water equipment. This cell, incidentally, is the only one of four cells which was not plastered and given a waterproof coating over the unmortered concrete block of the shield. The section of the oxygen line in the heavy water cell at the time of the initial leak was provided with two valves: one an air operated valve to admit oxygen, and the other a thermal expansion valve for regulating the flow of gas to a few cubic centimeters per minute. The oxygen line continues into the low pressure fuel cell through a surge check valve and into an oxygenation chamber containing fuel solution, normally at 1000 psi and 60-75° C.

The cause of the first leak was a difficulty in the thermal expansion valve which resulted in a low oxygen flow. After attempts to free the valve by increasing its temperature (the valve was electrically heated) and by shock, which may have weakened the welded joint upstream of the valve, the weld failed and an undetermined amount of fuel solution was discharged into the heavy water cell. The check valve upstream of the valve prevented further loss before the fuel could be dumped. Based on a visual examination at that time, the loss was estimated at less than 5 liters of solution containing 30 grams of U^{235}/liter. Subsequently, the walls of the cell were flushed with water to the waste evaporator system and the thermal expansion valve was replaced by an air operated regulating valve. This leak was not considered serious enough to suspend reactor operation and experiments were continued.

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DATE 4/14/67
For The Atomic Energy Commission
Chief, Declassification Branch

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X-81 (6-51)

Five days later a leak was observed in a threaded connection of the high pressure check valve on the oxygen line. On the basis of the visible deposit of fuel solution in the vicinity of the leak, the loss was again judged to be small, although it was not known how long the leak had been present. Not until samples and volume measurements could be taken of the fuel solution remaining in the system was the possible magnitude of the two losses suspected. Again, clean-up of the cell was effected by flushing the contaminated portions with water which carried the spilled fuel to the waste recovery tank.

Soon afterward, the remaining fuel was removed from the reactor and an accurate balance was made. The discrepancy in the amounts of fuel originally added and removed was found to be 780 grams of U^{235}.

At the time it was expected that the missing material could be washed from the cells and would appear in the waste evaporator to which the shield drains. However, after washing the cells and concentrating the wash, only 336 grams of U^{235} could be found as 80% enriched uranium. A second wash and concentration yielded 75 grams of U^{235} at $\sim$ 50% enrichment. The third wash produced only 10 grams at 10% enrichment, and the total recovery from the waste system amount to 415 grams of the 780 grams lost.

In view of the unsatisfactory state of the recovery effort the system was examined for escape routes other than the waste evaporator. The several rather obvious possibilities were:

1. Accidental discharge of waste material from the waste system storage tank to the drainage ditch.

2. Loss into the shield ventilation system.

3. Leakage into the shielding structure or earth surrounding the reactor fuel piping.

4. Leakage from reactor piping into surrounding heating or cooling jackets and subsequent loss in a condensate or waste water stream.

5. Back leakage into oxygen gas cylinders.

6. Errors in inventory or bookkeeping with respect to additions to, and withdrawals from, system.

7. Theft of the material.

The measures undertaken for investigation of each of these possibilities will be described.

<u>Accidental discharge of waste material from the waste evaporator system.</u>

The piping arrangement at the waste evaporator is such that the storage tank can be jetted to the waste water ditch by opening a valve which by-passes the waste evaporator. So far as can be established, the by-pass valve has not been opened since the final clean-out previous to filling the reactor with enriched fuel. Excessive entrainment from the evaporator during concentration of material in the evaporator also may have been a source of loss, although each batch of condensate was required to analyse less than one ppm uranium before discharging to the sewer. However, to obtain more substantial evidence, the discharge piping and the drainage ditch were carefully examined for evidence of uranium. The area was surveyed with both α, and β, γ detectors with no indication of activity. Smear samples taken from inside the discharge pipe and from algae growth on the tile-lined drainage ditch showed no alpha activity. Mud samples collected at several points downstream indicated a trace of uranium which could not be found on re-sampling.

<u>Loss into the shield ventilation system.</u> Cooling air flows through the shield continuously at a rate of $\sim$2500 cfm. The velocity of air past the leak point was approximately 25 ft./min. Judging from the large area of contamination in the low pressure fuel cell, the leak from the check valve was in the form of a fine spray. The air temperature at the time of the leak was approximately 120° F and the humidity, even assuming 100% for the cool incoming air, must have been less than 20%. These conditions suggest that drying of the mist particles and subsequent removal by the air current might be expected and, in fact, if one assumes that the leak continued as a spray over a period of a few hours, it can be shown that 20 or 30 liters of fuel solution could be evaporated to dryness and removed by the ventilation system.

A survey was made of the cooling air duct from the point at which it leaves the reactor cell to the top of the ventilating stack. There was indication of considerable alpha activity throughout the duct work, and smear samples taken at several points indicated the presence of enriched uranium.

Although there is no good way of estimating what percentage of particulate matter might be picked up by the duct walls, samples taken from 1 sq. ft. of area were used to estimate that a maximum of 2 or 3 grams might have stuck to the duct. Based on this, one would have to assume a removal efficiency of 0.5% to account for the missing uranium.

The records of the Laboratory meteorological group show that a wind of 1 to 4 miles/hr. was blowing in a westerly direction during the period in which the loss from the check valve is likely to have occurred. One can estimate that the plume of gas might touch ground level at a distance of 1500 ft. from the stack and that the concentration at this point would be $\sim 10^{-7}$g of uranium/cu.ft. of air.

Assuming that the material had been dispersed from the stack, an alpha activity survey was made of the area surrounding the stack starting on the roof of Bldg. 7500 and following the path of the prevailing winds for several hundred yards up and down the valley. Although no indication of alpha activity was discovered, this is not particularly surprising because of the high dilution expected in the dispersing stack gases and when one considers the relatively low sensitivity of any α - survey instruments.

In spite of the fact that we have been unable to find very conclusive evidence that the loss was by this route, it is believed that this is the best of the possibilities which have been investigated.

Leakage into the shielding structure or earth surrounding the reactor fuel piping. When it was learned that all of the lost material had not been recovered from the waste evaporator system, a program was begun to locate any reactor piping leaks which might be present, but not visible. After repairing the known leaks, the system was completely filled with water and hydrostatic pressure of 400 psi applied to both the high and low pressure piping simultaneously. All lines leading outside the shielding equipment were cut and welded closed, to be certain that leakage into the liquid or gaseous waste piping would not be confused with leaks from the fuel system.

A small leak was found past the gasket of the flame recombiner in the low pressure system. This joint must normally withstand a few pounds pressure and is not normally in contact with uranium solution.

Since there was no evidence in the form of dried uranyl sulfate and no detectable contamination, it was assumed that no soup leak had occurred from the flame recombiner. From the results of hydrostatic tests it was concluded that any significant loss by leakage from reactor piping must have been from the two original leaks.

An inspection of the cells in which the leaks occurred showed considerable yellow coloration from uranyl sulfate which had dried on the masonry walls and piping after having been flushed with water. Measures were taken to remove all traces of contamination by scrubbing the piping and chipping away the wall surfaces. This task required the efforts of ten people over a period of two weeks, and unfortunately yielded only 10 grams of uranium in 30-40# of concrete. It is known that some material which soaked into the cracks of the loosely stacked block remains unrecovered, but it is believed that the quantity remaining is probably not greater than the 10 grams already recovered.

Leakage from reactor piping into surrounding heating or cooling jackets and subsequent loss in a condensate or waste water stream. Several pieces of reactor equipment containing fuel are surrounded by jackets which, although pressure tested previously, might cover leaks in fuel piping. During the hydrostatic pressure tests all of the jackets which normally contain steam or cooling water were given a pressure test. In no case was it possible to find a leak in any of these places. Furthermore, where possible, samples were taken from the fluids which were normally circulated through these and other jackets, e.g., soup pump cooling oil, 10 and 40 KW boiler feed water, the convection cooling system on the jackets which surround the fuel dump tanks, and from the heavy water which circulates outside the reactor core. In no case was there evidence that either uranium or sulfate ions were present in these fluids. Mud samples were taken from the steam condensate waste ditch and produced only negative results when examined for uranium. It was concluded that the uranium loss had not been through any jackets surrounding fuel piping.

Back leakage into oxygen gas cylinders. At the time of the difficulty with the oxygen regulating valve, as described in the introduction of this memo, the flow of oxygen was discontinuous and made it possible, though unlikely, that uranium solution backed up past the check valve, regulating valve and shut-off valve, through the oxygen pressure reducing system and into the supply cylinders. This would have required simultaneous leakage of several valves and the unlikely situation of a lower pressure in the O_2 cylinder than in the system. The oxygen line was dismantled and flushed, first from the reactor shield to the regulating station in the control room, and then from the regulating station back to the oxygen cylinder connection. Samples of these washes indicated 0.2 gram removed from the reactor side of the line but only 0.02 gram total from the cylinder side of the line. It is known that the contamination could have resulted from an earlier incident in which the oxygen pressure on the reactor was intentionally vented at a relief valve in the control room to serve the purpose of a particular experiment. It was noted at the time, which incidentally was several weeks previous to the spillage accident, that droplets of uranium solution had escaped from the vent point. Although the vent point was on the reactor side of the regulator it is reasonable to expect that traces of uranium solution might have been forced past the regulator at times when oxygen cylinders were being changed, which would account for the presence of uranyl sulfate on both sides of the vent line. Should 5 or 10 liters of the highly acid sulfate have entered the cylinders it is highly probable that the brass cylinder valve or the cylinder itself would have been damaged or that the presence of the liquid would have been noted by the cylinder company or by some person at ORNL (all ORNL cylinders are returned to ORNL after having been refilled). The cylinder gas company and the Laboratory Stores group were contacted and neither had information which suggested a clue. The brass reducer station at the HRE was examined for evidence of corrosion which might have resulted from contact with fuel solution but no evidence was found. Further investigation of this loss possibility was discontinued.

Errors in inventory or bookkeeping with respect to additions to, and withdrawals from system. Considering that an error may have been made in the quantities of materials added to, or withdrawn from the system, a re-examination was made of all the detailed bookkeeping records covering the period of operation with enriched material. These records were checked independently and found to verify the quantities involved in each transaction. It is believed that the loss indicated by the records is accurate to at least ±25 grams, this being determined by analytical and weighing error.

Theft of material. In our opinion the possibility that the material may have been stolen can be ruled out for several reasons. Although it is somewhat beyond the scope of the Operations group to pursue such a matter, there appear to be no suspicious circumstances or unusual incidents which might be related to the loss. The integrity of the personnel having access to the material is unquestioned by us, and it is doubtful that the withdrawal of solution from the system could have been accomplished by a person unfamiliar with the reactor. Furthermore, the practical difficulties attendant to removal of a large volume ($\sim$ 20 liters) of liquid from the restricted 7500 Area, without attracting the attention of either the Operating or Security personnel, are considerable. At no time since receipt of the fissionable material has the double-fenced area been unguarded.

Summary

Approximately six weeks have been spent in investigating these various possibilities. The loss to the shield ventilating system remains as the most reasonable explanation. The present accounting based on final assays and analyses of all material so far recovered shows: Spilled - 780.06; Recovered - 411.37; Unrecovered - 368.69.

Measures have been taken to prevent repetition of such a loss from the reactor when it is operated in the future. First the beta, gamma activity of the fuel solution will be increased to a detectable level by raising the reactor to higher power levels for a short time in order to make use of the activity monitors designed into the system to warn of leakage. Furthermore, continuous monitoring of the stack gases with a precipitron type sampling device to be installed will indicate the presence of alpha activity, as well as beta and gamma activity. The hydrostatic leak detector system attached to piping flanges has been overhauled completely, although neither of the leaks covered in this memo would have been detected by this device. In addition, more emphasis will be placed on frequent inventories from samples and the instruments indicating the quantity of liquid in the system, although it is unlikely that a loss of less than 300 grams will be obvious from such an inventory.

S E Beall

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SEB:dk